

On two-phonon processes in neutron diffraction against crystals

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With 5 figures in the text

ABSTRACT

The two-phonon processes in slow neutron scattering against single crystals are discussed. It is shown that in the harmonic approximation the energy distribution of neutrons scattered coherently in a certain direction in some cases has logarithmic singularities for the energies obtained in the one-phonon processes. Further there appear singularities of the same kind as Van Hove found earlier for the incoherent one-phonon scattering. To illustrate the logarithmic singularities more explicit results are given for an ideal Debye crystal. A formula for the incoherent two-phonon scattering is also given, and this is applied on KCl.

I. Introduction

In recent years several successful experiments have been performed to determine the eigen vibrations of crystals, using scattering of slow neutrons [1-5]. One there uses the fact that in processes where one vibrational quantum participates the neutrons are scattered with a certain number of discrete energies, which are well separated and can thus be measured. Such experiments will certainly be a valuable complement to those on diffuse X-ray scattering [6-9] in providing more complete information about the crystal vibrations. So far much stronger intensities are obtained using X-rays, but owing to the fact that the vibrational frequencies are determined from the measured scattering intensities, corrected for Compton scattering, higher phonon scattering etc., the interpretation of the experimental results is not as direct as for neutrons. When using neutrons only the scattering energies are measured, and for a well monochromized incident beam the intensities mostly have sharp peaks for energies, corresponding to one-phonon scattering. Owing to incomplete monochromatization and collimation the peaks always have finite widths.¹ We cannot *a priori* assume that especially the coherent two-phonon scattering does not display more or less pronounced intensity maxima and minima.² If such exist they may in some cases confuse the interpretation of the experimental results. It should also be noted that the incoherent one-phonon scattering may have a similar confusing effect.³

¹ Van Hove has shown that even under ideal experimental conditions the one-phonon scattering surfaces have finite widths, owing to the anharmonicity of the crystal vibrations.

² Brockhouse and Stewart [2] claimed to have found indications of such maxima in their experiments. Carter, Palevsky and Hughes [4] also got some peaks that could not be interpreted as one-phonon peaks, but as Palevsky informed the author these results were very uncertain.

³ See G. Placzek and L. Van Hove, Phys. Rev. 93, 1207 (1954).

So far very few discussions of the corrections to the one-phonon scattering due to higher phonon processes have been published.¹ General expressions for their intensities are given by Fröman [10], Glauber [11] and Van Hove [12].² It is, however, hard to draw any further conclusions from their formulas without a more detailed analysis of these expressions. Several authors [13–16] have discussed the effect of the higher phonon processes on the total cross section, but here we are interested in the differential scattering cross section, indicating the energy distribution of the scattered neutrons. Similar problems arise in connection with X-ray diffraction, but owing to the fact that different cross sections are considered in the two cases the results on X-rays cannot be applied to neutrons [6–9].

In the one-phonon processes the energies of the scattered neutrons are correlated to the eigen frequencies of the crystal by the following energy-momentum relation [17]:

$$\left. \begin{aligned} k^2 &= k_0^2 - \varepsilon \frac{2m}{\hbar} \omega_j(q), \\ \mathbf{k} &= \mathbf{k}_0 + \mathbf{q} - \boldsymbol{\tau}. \end{aligned} \right\} \quad (1)$$

$\mathbf{k}_0$, $\mathbf{k}$ and $\mathbf{q}$ are the wave vectors of the incident and scattered neutron and the wave vector of the eigen vibration, participating in the scattering. $\boldsymbol{\tau}$ is an arbitrary reciprocal lattice vector, defined in such a way that the product of this and a direct lattice vector is a multiple of 2π . $\omega_j(\mathbf{q})$ denotes a certain branch of the eigen frequencies and m is the neutron mass. ε can take the values ± 1 , corresponding to phonon absorption and phonon emission respectively.

In the following we will discuss the two-phonon processes to determine the characteristic features of the scattering. Our main aim is to ascertain whether the cross section shows any singularities and to get an idea how sensitive it is to the details of the crystal vibrations.

II. The high phonon processes

For the low order phonon processes we know that the coherent and incoherent scattering cross sections vary in very different ways with the scattering angle and energy, the coherent ones showing stronger singularities. Concerning high phonon processes it can, however, be shown that the two kinds of cross section are approximately proportional. The intensity from the n -phonon processes is approximately given by³

$$\frac{d^2 \sigma^{(n)}}{d\Omega d\omega} \approx N a^2 \frac{k}{k_0} e^{-2W} \frac{(2W)^n}{n! \sigma_0 (2\pi n)^{\frac{1}{2}}} \phi_n(\omega - \omega_0), \quad n \geq 1. \quad (2)$$

N is the number of nuclei in the crystal. a should be set equal to the coherent or incoherent scattering length respectively, and e^{-2W} denotes the ordinary Debye-Waller factor. $\hbar\omega_0$ and $\hbar\omega$ are the energies of the incident and scattered neutron

¹ Some results are given by P. A. Egelstaff in Harwell Reports AERE-1164. Some remarks may also be found in: G. Plazcek and L. Van Hove, loc. cit.

² The formal treatment goes back to ideas found in A. Akhiezer and I. Pomeranchuk, J. Exptl. Theoret. Phys. U.S.S.R., 17, 770 (1947), though these authors give explicit formulas only in the incoherent approximation.

³ The proof will be published in a later paper.

and $\frac{1}{\sigma_0 (2\pi n)^{\frac{1}{2}}} \phi_n(\omega - \omega_0)$ is a normalized Gaussian function with the width $\sigma_0 \sqrt{n}$ and the maximum value at $\omega - \omega_0 = -nm$, σ_0 and m being temperature dependent constants independent of n . From (2) we thus conclude that when $2W < 1$ the intensities from the higher phonon terms decrease rapidly with increasing order. We can also assume that the higher order scattering varies much more smoothly than for instance the second order scattering. In typical one-phonon measurements using cold neutrons even the two-phonon intensities are rather weak, and we may therefore neglect all higher phonon terms.

III. The coherent two-phonon processes

For brevity a Bravais' lattice only will be considered. As a starting point we may take the following expression for the coherent two-phonon cross section per unit cell, given by Fröman [10]:¹

$$\frac{d^2 \sigma_{\text{coh}}^{(2)}}{d\Omega d\omega} = a_{\text{coh}}^2 \frac{(2\pi)^3 k}{V k_0} e^{-2W} \frac{1}{2} \sum_{s, \varepsilon} \sum_{s', \varepsilon'} \frac{\hbar (\mathbf{x} \cdot \mathbf{C}_s)^2}{2NM\omega_s} \frac{\hbar (\mathbf{x} \cdot \mathbf{C}_{s'})^2}{2NM\omega_{s'}} \times \\ \times \frac{\coth(\beta\omega_s) + \varepsilon}{2} \frac{\coth(\beta\omega_{s'}) + \varepsilon'}{2} \delta_P(\mathbf{x} + \mathbf{q} + \mathbf{q}') \delta(\omega - \omega_0 + \varepsilon\omega_s + \varepsilon'\omega_{s'}). \quad (3)$$

Some of the notations have been explained above and the new ones denote the following:

$\mathbf{C}_s$ = polarization vector of an eigen vibration, characterized by the double index $s = (\mathbf{q}, j)$.²

M = mass of a nucleus.

V = volume of the elementary cell of the crystal.

$\beta = \hbar/2k_B T$, where k_B = Boltzmann's constant, T = temperature.

$\mathbf{x} = \mathbf{k}_0 - \mathbf{k}$.

$\delta(\dots)$ is an ordinary Dirac function and $\delta_P(\mathbf{x}) = \sum_{\boldsymbol{\tau}} \delta(\mathbf{x} - \boldsymbol{\tau})$ denotes a periodic Dirac function singular at the reciprocal lattice points. A double summation should be performed over the eigen states $s = (\mathbf{q}, j)$ and $s' = (\mathbf{q}', j')$ and a summation should also be made over the two values ± 1 for ε and ε' .

The Dirac functions express the conservation of energy and momentum, analogous to the case of the one-phonon scattering (see Fig. 1).

$$\left. \begin{aligned} k^2 &= k_0^2 - \varepsilon \frac{2m}{\hbar} w_j(\mathbf{q}) - \varepsilon' \frac{2m}{\hbar} w_{j'}(\mathbf{q}'), \\ \mathbf{k} &= \mathbf{k}_0 + \mathbf{q} + \mathbf{q}' - \boldsymbol{\tau}. \end{aligned} \right\} \quad (4)$$

We note that two phonons are now involved.

¹ The notations used here are not exactly the same as in Fröman's paper.

² $\mathbf{C}_s$ is normalized according to $|\mathbf{C}_s| = 1$.

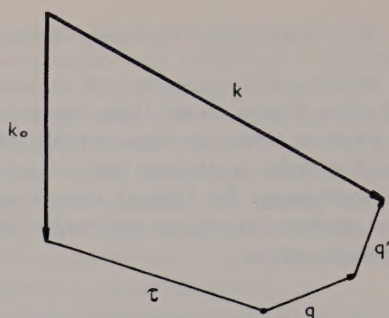


Fig. 1.

Before any conclusions are drawn from the above intensity formula, the latter will be brought into a more convenient form. To save space we introduce the notation

$$f_j(\mathbf{q}, \varepsilon) = \frac{(\mathbf{e}_{\mathbf{x}} \cdot \mathbf{C}_s)^2}{2\omega_s} [\coth(\beta\omega_s) + \varepsilon]; \quad (5)$$

$\mathbf{e}_{\mathbf{x}}$ then means the unit vector along $\mathbf{x}$.

The summations over $\mathbf{q}$ and $\mathbf{q}'$ are transformed into integrations over the wave vectors as

$$\sum_{\mathbf{q}} \rightarrow \frac{NV}{(2\pi)^3} \int_{(\text{cell})} \dots d\mathbf{q}$$

and new integration variables $\mathbf{q}_1$ and $\mathbf{q}_2$ are introduced as follows:

$$\mathbf{q}_1 = \mathbf{q}, \quad \mathbf{q}_2 = \mathbf{q} + \mathbf{q}'.$$

The δ_P -function only contributes when $(\mathbf{x} + \mathbf{q}_2)$ reaches a reciprocal lattice point and after the $\mathbf{q}_2$ -integration we can write

$$\begin{aligned} \frac{d^2 \sigma_{\text{coh}}^{(2)}}{d\Omega d\omega} &= a_{\text{coh}}^2 \frac{k}{k_0} e^{-2W} \frac{1}{2} \left(\frac{\hbar \kappa^2}{2M} \right)^2 \frac{V}{(2\pi)^3} \sum_{j,j'} \sum_{\varepsilon, \varepsilon'} \sum_{\tau} \times \\ &\times \int_{V_{\tau}} f_j(\mathbf{q}_1, \varepsilon) f_{j'}(\boldsymbol{\tau} - \mathbf{x} - \mathbf{q}_1, \varepsilon') \delta[\omega - \omega_0 + \varepsilon\omega_j(\mathbf{q}_1) + \varepsilon'\omega_{j'}(\boldsymbol{\tau} - \mathbf{x} - \mathbf{q}_1)] d\mathbf{q}_1. \end{aligned}$$

$\sum_{\tau}$ denotes the summation over the eight possible lattice points above, and the volumes of integration are determined from the fact that both $\mathbf{q}_1$ and $(\mathbf{q}_2 - \mathbf{q}_1)$ should be contained in a reciprocal unit cell. The volumes V_{τ} do not overlap and taken together they precisely fill a unit cell. Using the periodicity of $f_j(\mathbf{q}, \varepsilon)$ and also noting that $f_j(-\mathbf{q}, \varepsilon) = f_j(\mathbf{q}, \varepsilon)$ we can simplify our expressions as follows:¹

$$\sum_{\tau} \int_{V_{\tau}} \dots d\mathbf{q}_1 = \int_{(\text{cell})} f_j(\mathbf{q}_1, \varepsilon) f_{j'}(\mathbf{x} + \mathbf{q}_1, \varepsilon') \delta[\omega - \omega_0 + \varepsilon\omega_j(\mathbf{q}_1) + \varepsilon'\omega_{j'}(\mathbf{x} + \mathbf{q}_1)] d\mathbf{q}_1.$$

¹ This latter transformation is used only to get a more attractive form.

The δ -function stipulates that only those $\mathbf{q}_1$ -vectors should be considered which lie on certain surfaces in the unit cell, defined by:

$$\lambda_{jj', \varepsilon \varepsilon'} = \omega - \omega_0 + \varepsilon \omega_j(\mathbf{q}) + \varepsilon' \omega_{j'}(\mathbf{x} + \mathbf{q}) = 0. \quad (6)$$

Let us call such a surface a λ -surface. We note that we get one λ -surface for each combination of the branches jj' and also for each combination of $\varepsilon \varepsilon'$. Further the λ -surfaces vary with $\mathbf{x}$, a fact which makes the numerical calculations very tedious when the intensities for many $\mathbf{x}$ -vectors are required.

The above δ -function can be eliminated by transforming to a surface integral. Thus denoting a λ -surface by S_λ and with $\text{grad } \lambda$ meaning the gradient with respect to $\mathbf{q}$ we may write^{1, 2}

$$\begin{aligned} \frac{d^2 \sigma_{\text{coh}}^{(2)}}{d\Omega d\omega} &= a_{\text{coh}}^2 \frac{k}{k_0} e^{-2W} \frac{1}{2} \left(\frac{\hbar \kappa^2}{2M} \right)^2 G_2^{\text{coh}}(\mathbf{x}), \\ G_2^{\text{coh}}(\mathbf{x}) &= \frac{V}{(2\pi)^3} \sum_{jj', \varepsilon \varepsilon'} \int_{S_\lambda} \frac{f_j(\mathbf{q}, \varepsilon) f_{j'}(\mathbf{x} + \mathbf{q}, \varepsilon')}{|\text{grad } \lambda|} dS. \end{aligned} \quad (7)$$

When determining the possible singularities of the cross section we must pay special attention to the following critical points of the integrals:

- (1) $\mathbf{q} = 0$, $\mathbf{q} = \boldsymbol{\tau} - \mathbf{x}$, in which points $f_j(\mathbf{q}, \varepsilon)$ or $f_{j'}(\mathbf{x} + \mathbf{q}, \varepsilon')$ are infinite.
- (2) zero-points of $\text{grad } \lambda$.

The intensity formula shows some characteristic similarities to the corresponding expression for the incoherent one-phonon scattering [18].³

$$\left. \begin{aligned} \frac{d^2 \sigma_{\text{incoh}}^{(1)}}{d\Omega d\omega} &= a_{\text{incoh}}^2 \frac{k}{k_0} e^{-2W} \frac{\hbar \kappa^2}{2M} G_1^{\text{incoh}}(\omega - \omega_0), \\ G_1^{\text{incoh}}(\omega) &= \frac{f(\omega)}{2\omega} [\coth(\beta\omega) - 1], \quad |\omega| \leq 1, \\ &= 0, \quad |\omega| > 1. \end{aligned} \right\} \quad (8)$$

$f(\omega)$ means the vibrational frequency function with the neutron "frequency" ω inserted instead of the vibrational frequency, and $f(\omega)$ is defined for negative frequencies as follows:

$$f(-\omega) = f(\omega)$$

and normalized to unity⁴

$$\int_0^1 f(\omega) d\omega = 1.$$

¹ In S_λ and $\text{grad } \lambda$ we have for brevity omitted the indices $jj' \varepsilon \varepsilon'$.

² It must be noted that for cold neutrons the λ -surfaces corresponding to emission of one or two phonons do not always exist, which implies that the corresponding integrals vanish.

³ The formula is exactly valid only for simple crystals with cubic symmetry. See also G. Placzek and L. Van Hove, loc. cit.

⁴ The frequencies are measured in units of the maximum vibrational frequency.

To make the resemblance more obvious $G_1^{\text{incoh}}(\omega - \omega_0)$ is written in the following altered form:

$$G_1^{\text{incoh}}(\omega - \omega_0) = \frac{V}{3(2\pi)^3} \sum_{j, \varepsilon} \int_{S_\lambda} \frac{g_j(\mathbf{q}, \varepsilon)}{|\text{grad } \lambda|} dS, \quad (9)$$

where
$$g_j(\mathbf{q}, \varepsilon) = \frac{1}{2\omega_s} [\coth(\beta\omega_s) + \varepsilon]. \quad (9')$$

The integration should now be performed on the iso-frequency surfaces

$$\lambda_{j, \varepsilon} = \omega - \omega_0 + \varepsilon\omega_j(\mathbf{q}) = 0. \quad (9'')$$

We know that the incoherent one-phonon cross section varies continuously and that singularities of the following type appear [19]:

$$\begin{aligned} \frac{d^2 \sigma_{\text{incoh}}^{(1)}}{d\Omega d\omega} &= \text{const } |\omega - \omega_c|^{\frac{1}{2}}, & \omega > \omega_c, \\ &= 0, & \omega < \omega_c. \end{aligned} \quad (9''')$$

These are due to the vanishing of $\text{grad } \lambda$. Moreover for some crystals the intensity shows sharp maxima and minima.¹

Owing to the resemblance of (7) and (9) we might draw similar conclusions for the coherent two-phonon cross section. To ascertain the existence or non-existence of singularities in the two-phonon intensity let us consider the critical points in more detail.

IV. The critical points $\mathbf{q} = 0$ and $\mathbf{q} = \boldsymbol{\tau} - \boldsymbol{\kappa}$

From (6) we see that when a λ -surface passes through either $\mathbf{q} = 0$ or $\mathbf{q} = \boldsymbol{\tau} - \boldsymbol{\kappa}$, where $\boldsymbol{\tau}$ is chosen so that $\mathbf{q}$ lies within the reciprocal unit cell, the momentum vector and the energy of the scattered neutron are correlated in accordance with the one-phonon scattering conditions. Let us consider a certain branch of the one-phonon scattering surfaces, which thus is determined from

$$\omega - \omega_0 + \varepsilon_1\omega_{j_1}(\boldsymbol{\kappa}) = 0$$

for definite ε_1 and j_1 . When this condition is fulfilled the λ -surfaces indicated by

$$(jj', \varepsilon \varepsilon') = (j_1 j_1, \varepsilon_1 \varepsilon_1)$$

for arbitrary j and ε go through $\mathbf{q} = 0$. Correspondingly the λ -surfaces with the indices

$$(jj', \varepsilon \varepsilon') = (j_1 j_1', \varepsilon_1 \varepsilon_1')$$

for arbitrary j' and ε' go through the point $\mathbf{q} = \boldsymbol{\tau} - \boldsymbol{\kappa}$.

The two critical points are completely equivalent and therefore we need only consider one of them, say $\mathbf{q} = 0$. To determine how a corresponding integral included

¹ See for instance the frequency spectra drawn by Jakobsen [8] for Cu and by Walker [9] and Squires [20] for Al.

in (7) varies when $\mathbf{x}$ approaches a point $\mathbf{x}_c$ on the one-phonon scattering surface, $\mathbf{x}$ is supposed near $\mathbf{x}_c$ outside the surface (Fig. 2). In that case the λ -surface is approximately determined from the equation below, if we keep near $\mathbf{q} = 0$.

$$\lambda = \varepsilon c_j(\mathbf{q}) |\mathbf{q}| + \varepsilon' \text{grad } \omega_j(\mathbf{x}_c) \cdot \mathbf{q} - \mu = 0, \quad (10)$$

where $c_j(\mathbf{q})$ denotes the sound velocity. μ is defined by

$$\mu = [\mathbf{v} - \varepsilon' \text{grad } \omega_j(\mathbf{x}_c)] \cdot (\mathbf{x} - \mathbf{x}_c), \quad (10')$$

where $\mathbf{v}$ denotes the velocity vector of the scattered neutron.

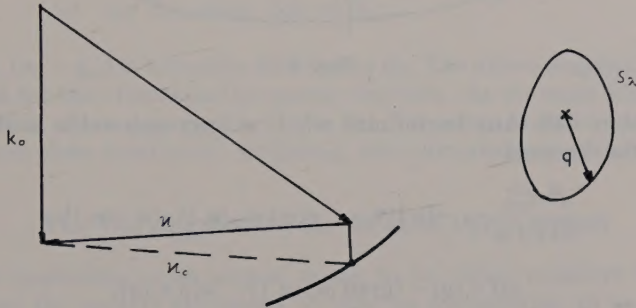


Fig. 2.

Suppose that $\text{grad } \lambda \neq 0$ everywhere on the λ -surface. Otherwise we would have an even stronger singularity. Except for a strictly isotropic crystal c_j depends on the direction of $\mathbf{q}$, and we may have the following two cases:

1. $|\text{grad } \omega_j(\mathbf{x}_c)| < \inf c_j(\mathbf{q})$:

The integration surface is contained within a sphere with radius $r \approx \mu$, and we have mainly to evaluate the integral

$$\int_{S_\lambda} \frac{dS}{[\omega_j(\mathbf{q})]^2}$$

which tends to a finite value, when $\mu \rightarrow 0$. We can also show that the intensity tends to the same value when approaching the one-phonon surface from both sides. This case will surely occur, for $\text{grad } \omega_j(\mathbf{x}_c)$ actually vanishes at certain points, and for j denoting the longitudinal and j' a transversal branch c_j will ordinarily be larger than $|\text{grad } \omega_j|$ that is when the one-phonon surface corresponds to transversal vibrations.

2. $|\text{grad } \omega_j(\mathbf{x}_c)| > \sup c_j(\mathbf{q})$:

The λ -surface has the shape indicated in Fig. 3 and we may proceed as follows, excluding all constant factors and only considering the order of magnitude:¹

$$\int_{S_\lambda} f_j(\mathbf{q}, \varepsilon) f_{j'}(\mathbf{x} + \mathbf{q}, \varepsilon') \frac{dS}{|\text{grad } \lambda|} \sim \int_{S_\lambda} \frac{dS}{|\mathbf{q}|^2} \sim \int_0^{(\infty)} \frac{r dr}{r^2 + \mu^2} \sim \ln \left| \frac{1}{\mu} \right|.$$

¹ r is inserted in the figure as the radius, using cylindrical coordinates.

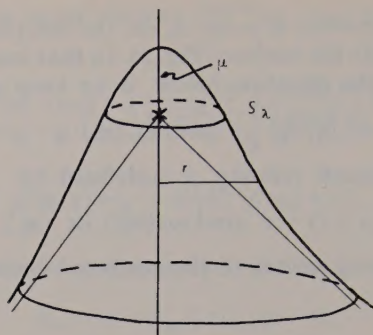


Fig. 3.

The cross section will thus be infinite when $\mathbf{x}$ approaches the scattering surface according to the formula

$$\frac{d^2 \sigma_{\text{coh}}^{(2)}}{d\Omega d\omega} \sim -\ln \left| [\mathbf{v} - \varepsilon' \text{grad } \omega_{j'}(\mathbf{x}_c)] \cdot (\mathbf{x} - \mathbf{x}_c) \right|. \quad (11)$$

When $\inf c_j(\mathbf{q}) < |\text{grad } \omega_{j'}(\mathbf{x}_c)| < \sup c_j(\mathbf{q})$,

we may also have the same type of singularity, but the exact condition for this is rather complicated and will not be given here. This case may especially happen when j' refers to the longitudinal and j to a transversal branch or in other words if the one-phonon surface refers to longitudinal vibrations. Experimentally the singularity would appear as a broadening of the one-phonon scattering surface.

Considering the above derivation in more detail we may conclude that for scattering points approaching a reciprocal lattice point $\boldsymbol{\tau}$ along a one-phonon surface the intensity increases as $1/|\mathbf{x} - \boldsymbol{\tau}|^2$ besides the logarithmic increase above.

V. The critical points $\text{grad } \lambda = 0$

The zeros of $\text{grad } \lambda$ are critical points of our integrals only when they lie on the integration surfaces, and the conditions for this ($\lambda = \text{grad } \lambda = 0$) may be written in the following form:

$$\left. \begin{aligned} \varepsilon \omega_j(\mathbf{q}) + \varepsilon' \omega_{j'}(\mathbf{x} + \mathbf{q}) + \omega - \omega_0 &= 0, \\ \varepsilon \text{grad } \omega_j(\mathbf{q}) + \varepsilon' \text{grad } \omega_{j'}(\mathbf{x} + \mathbf{q}) &= 0. \end{aligned} \right\} \quad (12)$$

These equations determine a certain surface $S_{\mathbf{x}}$ with the following property:

For a $\mathbf{x} = \mathbf{x}_c$ on $S_{\mathbf{x}}$ there exists at least one critical point $\mathbf{q}_c$ of the corresponding integral.

A $\mathbf{x}$ near $S_{\mathbf{x}}$ is now considered and the contribution of the integral from the region near a critical point is estimated. Supposing the critical point to be analytic the λ -surface may be approximated by a surface of second degree, when keeping near $\mathbf{q}_c$. The equation of the surface can be written as

$$a_1 \xi_1^2 + a_2 \xi_2^2 + a_3 \xi_3^2 = \mu_1, \quad (13)$$

where $\xi = \mathbf{q} - \mathbf{q}_c$ and the coordinate axis have been chosen along the principal axis. a_1, a_2, a_3 are positive or negative constants and μ_1 is defined by

$$\mu_1 = [\mathbf{v} - \varepsilon' \text{grad } \omega_{j'}(\mathbf{x}_c + \mathbf{q}_c)] \cdot (\mathbf{x} - \mathbf{x}_c). \quad (14)$$

It is now a straightforward matter to evaluate the integral (see [21]), and this is shown to have a singularity of the following type, when crossing the $S_{\mathbf{x}}$ -surface:¹

$$\begin{aligned} \frac{d^2 \sigma_{\text{coh}}^{(2)}}{d\Omega d\omega} &= \text{const} \left| [\mathbf{v} - \varepsilon' \text{grad } \omega_{j'}(\mathbf{x}_c + \mathbf{q}_c)] \cdot (\mathbf{x} - \mathbf{x}_c) \right|^{\frac{1}{2}} \quad \text{on one side} \\ &= 0 \quad \text{on the other side of } S_{\mathbf{x}}. \end{aligned} \quad (15)$$

$[\mathbf{v} - \varepsilon' \text{grad } \omega_{j'}(\mathbf{x}_c + \mathbf{q}_c)]$ is normally directed to $S_{\mathbf{x}}$. The above singularity corresponds exactly to that for the vibrational frequency function. As the same kind of discussion may be found in other papers [19, 21] on the frequency function, we refer to these for a more complete treatment, including also non-analytic critical points.

VI. The singularities for an ideal Debye crystal

Though the scattering cross section seems to be rather sensitive to the detailed assumptions on the crystal vibrations, it may be illuminating to apply our results to an ideal Debye crystal. We then have

$$\omega_1 = \omega_2 = \omega_3 = \omega(\mathbf{q}) = c |\mathbf{q}| \quad (c = \text{sound velocity}),$$

if $\mathbf{q}$ is restricted to the reciprocal unit cell,² and the λ -surfaces defined in (6) are the same for all values of (j, j') . We can thus sum over these indices in formula (7) and write the result in the following form:

$$\sum_{j,j'} f_j(\mathbf{q}, \varepsilon) f_{j'}(\mathbf{q}', \varepsilon') = e^{\beta(\omega_0 - \omega)} \frac{1}{2\omega(\mathbf{q}) \sinh[\beta\omega(\mathbf{q})]} \frac{1}{2\omega(\mathbf{q}') \sinh[\beta\omega(\mathbf{q}')]}$$

with $\mathbf{q}' = \mathbf{x} + \mathbf{q}$.

For sufficiently high temperatures $\sinh[\beta\omega(\mathbf{q})]$ can be approximated by $1/\beta\omega(\mathbf{q})$ which further simplifies our expression. We get

$$G_2^{\text{coh}}(\mathbf{x}) = e^{\beta(\omega_0 - \omega)} \left(\frac{k_B T}{\hbar c^2} \right)^2 \frac{V}{(2\pi)^3} \sum_{\varepsilon, \varepsilon'} \int_{S_\lambda} \frac{1}{|\mathbf{q}|^2 |\mathbf{x} + \mathbf{q} - \boldsymbol{\tau}|^2} \frac{dS}{|\text{grad } \lambda|}, \quad (16)$$

where $\boldsymbol{\tau}$ is chosen so that $(\mathbf{x} + \mathbf{q} - \boldsymbol{\tau})$ always lies within the unit cell. The λ -surfaces obtained from

$$\varepsilon |\mathbf{q}| + \varepsilon' |\mathbf{x} + \mathbf{q} - \boldsymbol{\tau}| = \frac{\omega_0 - \omega}{c} \quad (17)$$

are either ellipsoids or hyperboloids or parts of these with their foci at $\mathbf{q} = 0$ and $\mathbf{q} = \boldsymbol{\tau} - \mathbf{x}$ depending on the position of the scattering point. We will especially deter-

¹ The continuous part of the cross section has been excluded in (15).

² In the Debye approximation the unit cell is spherical.

mine the scattering near the one-phonon scattering surfaces. Owing to the symmetry we need only consider one of these, for instance that corresponding to phonon emission ($\omega < \omega_0$). When we keep on one side of the surface $\omega_0 - \omega - c|\boldsymbol{\tau} - \boldsymbol{\kappa}| > 0$, and according to (17) only the integral, indicated by $(\varepsilon, \varepsilon') = (1, 1)$, contributes to the intensity. On the other side of the surface $\omega_0 - \omega - c|\boldsymbol{\tau} - \boldsymbol{\kappa}| < 0$, and in that case the non-vanishing integrals correspond to $(\varepsilon, \varepsilon') = (1, -1)$ and $(-1, 1)$. For a $\boldsymbol{\kappa}$ very near the scattering surface only those parts of the λ -surfaces passing near $\mathbf{q} = 0$ or $\mathbf{q} = \boldsymbol{\tau} - \boldsymbol{\kappa}^1$ need be considered, and the equations for them may be written in the form

$$\frac{x^2}{a_1^2} + \frac{y^2 + z^2}{b_1^2} = 1 \quad (18)$$

$$\text{with} \quad \left. \begin{aligned} a_1^2 &= \left(\frac{\omega_0 - \omega}{2c} \right)^2, \\ b_1^2 &= \left(\frac{\omega_0 - \omega}{2c} \right)^2 + \frac{|\boldsymbol{\tau} - \boldsymbol{\kappa}|^2}{4}. \end{aligned} \right\} \quad (18')$$

The x -axis has then been directed along the line between the two foci and the origin ($x = 0$) is placed midway between them. The following treatment will be separated into two cases as $b_1^2 \gtrless 0$.

$b_1^2 > 0$, corresponding to scattering points on one side of the one-phonon surface (Fig. 4a):

We may proceed as follows when evaluating the integral corresponding to $(\varepsilon, \varepsilon') = (1, 1)$. The others vanish.

$$\begin{aligned} \int_{S_\lambda} \frac{1}{|\mathbf{q}|^2 |\boldsymbol{\kappa} + \mathbf{q} - \boldsymbol{\tau}|^2} \frac{dS}{|\text{grad } \lambda|} &= 2\pi \int_{-a_1}^{a_1} \frac{r}{|\mathbf{q}|^2 |\boldsymbol{\kappa} + \mathbf{q} - \boldsymbol{\tau}|^2} \frac{dx}{|(\text{grad } \lambda)_n|} \\ &= \frac{2\pi}{\omega_0 - \omega} \int_{-a_1}^{a_1} \frac{dx}{|\mathbf{q}| |\boldsymbol{\kappa} + \mathbf{q} - \boldsymbol{\tau}|} \\ &= \frac{2\pi c}{(\omega_0 - \omega)^2} \int_{-a_1}^{a_1} \left[\frac{1}{|\mathbf{q}|} + \frac{1}{|\boldsymbol{\kappa} + \mathbf{q} - \boldsymbol{\tau}|} \right] dx \\ &= \frac{4\pi c}{(\omega_0 - \omega)^2} \int_{-a_1}^{a_1} \frac{dx}{|\mathbf{q}|}. \end{aligned}$$

$r = (y^2 + z^2)^{\frac{1}{2}}$ and $(\text{grad } \lambda)_n$ denotes the component of $\text{grad } \lambda$ perpendicular to the x -axis. We note that for q running along the λ -surface

$$|\mathbf{q}| = a_1 \left(1 + \frac{|\boldsymbol{\kappa} - \boldsymbol{\tau}|}{2a_1} \frac{x}{a_1} \right)$$

¹ $\boldsymbol{\tau}$ should then be chosen to restrict $(\boldsymbol{\tau} - \boldsymbol{\kappa})$ to the unit cell.

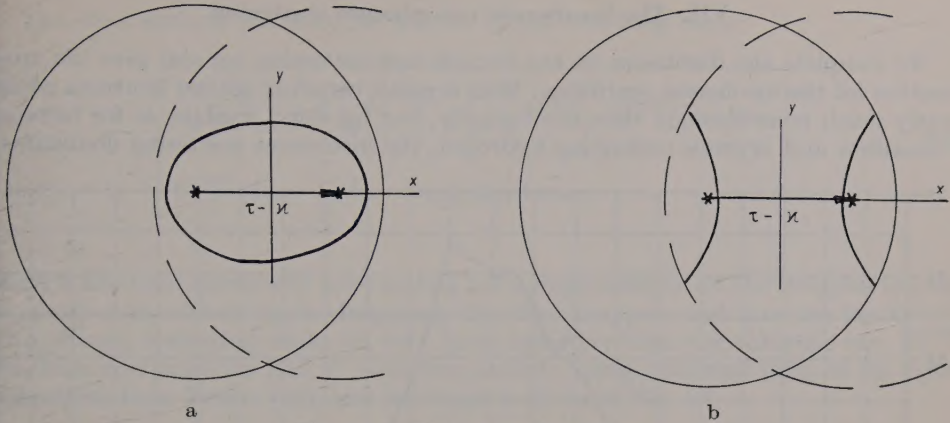


Fig. 4 a, b. The integration surfaces.

and the final integration thus gives the following result:

$$\int_{S_\lambda} \dots dS = \frac{4\pi}{|\tau - \kappa|(\omega_0 - \omega)} \ln \left| \frac{\omega_0 - \omega + c}{\omega_0 - \omega - c} \frac{|\tau - \kappa|}{|\tau - \kappa|} \right|. \quad (19)$$

$b_1^2 < 0$, corresponding to scattering points on the other side of the one-phonon surface (Fig. 4b):

We proceed in a similar way as above. Only the integrals corresponding to $(\varepsilon, \varepsilon') = (-1, 1), (1, -1)$ do not vanish. We now get

$$\sum_{\varepsilon, \varepsilon'} \int_{S_\lambda} \frac{1}{|q|^2 |\kappa + q - \tau|^2} \frac{dS}{|\text{grad } \lambda|} = \frac{4\pi c}{(\omega_0 - \omega)^2} \int_{a_1}^{(\infty)} \left[\frac{1}{|\kappa + q - \tau|} - \frac{1}{|q|} \right] dx$$

and further

$$|q| = a_1 \left(\frac{|\tau - \kappa|}{2a_1} \frac{x}{a_1} + 1 \right), \quad |\kappa + q - \tau| = a_1 \left(\frac{|\tau - \kappa|}{2a_1} \frac{x}{a_1} - 1 \right).$$

Extending the integration to infinity we obtain the same result as above.

Our formula will also be valid near the scattering surface for phonon emission if we only substitute $|\omega_0 - \omega|$ for $(\omega_0 - \omega)$.

The differential scattering cross section can now be written in the following form, valid in the Debye approximation and near a one-phonon scattering surface:^{1, 2}

$$\frac{d^2 \sigma_{\text{coh}}^{(2)}}{d\Omega d\omega} = a_{\text{coh}}^2 \frac{k}{k_0} \exp[-2W + \beta(\omega_0 - \omega)] \frac{1}{2} \left(\frac{\hbar^2 \kappa^2}{2Mk_B \theta} \frac{6T}{\theta} \right)^2 F(\kappa), \quad (20)$$

$$F(\kappa) = \frac{1}{12\omega_m} \frac{\omega_m^2}{\omega(\kappa)|\omega_0 - \omega|} \ln \left| \frac{|\omega_0 - \omega| + \omega(\kappa)}{|\omega_0 - \omega| - \omega(\kappa)} \right|.$$

Here $\theta =$ Debye temperature; $\omega_m = k_B \theta / \hbar =$ maximum vibrational frequency.

¹ Note that for cold neutrons the one-phonon surfaces corresponding to phonon emission disappear.

² Egelstaff (loc. cit.) considered this case only and his result essentially corresponds to formula (20), though he did not give this explicit expression.

VII. The incoherent two-phonon scattering

To complete the discussion on the two-phonon scattering we also give the cross section for the incoherent scattering. Most crystals certainly scatter neutrons coherently much more strongly than incoherently, but for some crystals, as for instance vanadium and crystals containing hydrogen, the incoherent scattering dominates.¹

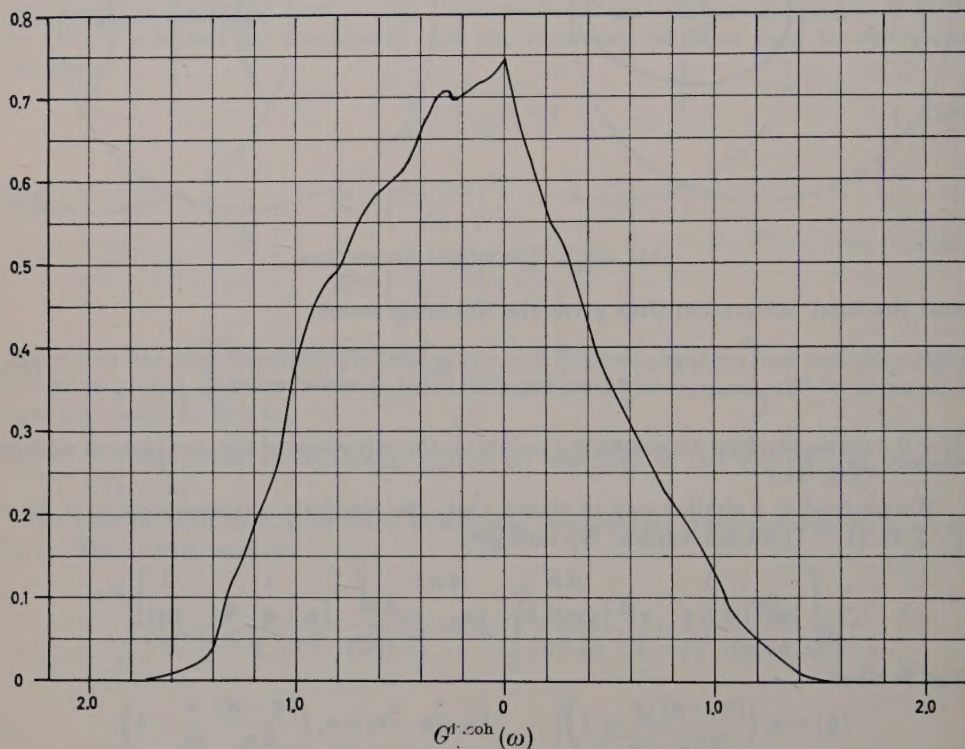


Fig. 5. To get the normalization proposed the values obtained from the figure must be multiplied by the factor 0.954.

The scattering cross section may be expressed in the vibrational frequency function $f(\omega)$, and for convenience we introduce the following notation:²

$$g(\omega) = \frac{f(\omega)}{2\omega\gamma(0)} [\coth(\beta\omega) - 1], \quad |\omega| \leq 1, \\ = 0, \quad |\omega| > 1, \quad (21)$$

where $\gamma(0)$ is defined from³ $\int_{-1}^1 g(\omega) d\omega = 1$.

¹ Recent experiments on vanadium have been made by Brockhouse [22], Carter, Hughes and Palevsky [23], and Stewart and Brockhouse [24]. Results from similar experiments on ZrH are found in refs. [26, 27].

² See formula (8) and the text below. The frequencies are measured in units of the maximum vibrational frequency. Compare also A. Sjölander, Ark. Fys. 13, 199 (1958).

³ Note that $2W = (\hbar\kappa^2/2M)\gamma(0)$.

The cross section can then be written in the following form:¹

$$\frac{d^2 \sigma_{\text{incoh}}^{(2)}}{d\Omega d\omega} = A \frac{k}{k_0} e^{-2W} \frac{1}{2} \left[\frac{\hbar \kappa^2}{2M} \gamma(0) \right]^2 G_2^{\text{incoh}}(\omega - \omega_0), \quad (22)$$

$$G_2^{\text{incoh}}(\omega) = \int_{-\infty}^{\infty} g(\omega - \omega_1) g(\omega_1) d\omega_1.$$

$4\pi A$ is the total incoherent cross section for a single nucleus. In the integration over ω_1 most of the details in the frequency function disappear and thus the incoherent two-phonon scattering ought to vary more smoothly than the coherent one. G_2^{incoh} has been computed for KCl at room temperature, using numerical data on the eigen vibrations from Tenerz [25], and the result is given in Fig. 5.²

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¹ The formula is exactly valid only for a simple crystal with cubic symmetry. A proof will be given in a later paper.

² The above formula is valid also for KCl, assuming the two ions to be equal apart from their charges. See A. Sjölander, loc. cit.

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